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The Trend of DDT and Parathion Residues on Apples Grown in Central Washington[1]

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Abstract

A study was carried out in 1938 to determine the average amounts of DDT and parathion residue on apples in central Washington and to compare these residues with those found in past years when fewer and earlier sprays and lower dosages were used. The DDT residue levels ranged from 0.3 to 12.4 p.p.m. with a mean of 3.0 p.p.m. for all samples, both washed and unwashed. For each variety and for all samples considered together, the unwashed apples had a higher DDT residue than the washed samples. Only nine of the unwashed samples (15.2%) and 1 washed sample (1.6%) had residues above the 7 p.p.m. tolerance level. Parathion residue ranged from 0.01 to 0.32 p.p.m. with a mean of 0.10 p.p.m. The residue values for both DDT and parathion reported here are somewhat higher than those reported for earlier years by various workers. With an increase in number of cover sprays applied, a corresponding increase in DDT residue was observed. Washing was found to lower the DDT residue level, but little difference between washing with and without brushes was apparent.

[1] From the Communicable Disease Center, Bureau of State Services, Public Health Service, U. S. Department of Health, Education and Welfare, Wenatchee, Washington. Accepted for publication April 24, 1939.

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Since the first general use of DDT and parathion for insect control on tree fruits in central Washington was made during 1947–48 the amount of these insecticides used each season to maintain adequate control has gradually increased. Although there has been a small increase in the dosage rate of DDT during this period, the major changes contributing to the residue problem have been the application of a greater number of cover sprays containing DDT during the growing season, and the fact that the last cover spray is often applied closer to harvest. The dosage rate and number of parathion sprays have also increased.

A large proportion of orchardists applied DDT sprays to their apples during 1947, and by 1948 practically all the apple orchards were sprayed with DDT. Parathion became available to orchardists during midseason of 1947, and by 1948 it was being widely used as a supplemental material in the spray program. In most instances only one or two DDT cover sprays were necessary for codling moth (*Carpocapsa pomonella* (L.)) control. Only a few orchards required more than three covers. During this period most orchardists used the dosages recommended by the State College of Washington (Anonymous 1948), *i.e.*, 1 pound of actual DDT per 100 gallons of water in the first cover spray, 0.5 to 1 pound in the second and third covers, the last cover to be applied not later than 30 days prior to harvest. In the second and third cover sprays a high percentage of orchardists used the 0.5-pound rather than the 1-pound DDT dosage. Parathion was recommended as a supplemental material where aphids and mites were a problem, at dosages of from 1 to 2 ounces of actual parathion per 100 gallons of water, with the spray not to be applied later than 30 days prior to harvest. During 1947 and 1948, parathion generally was used in only one or two cover sprays.

Westlake & Fahey (1950) analyzed samples of apples from the Yakima Valley area of Washington that were sprayed during the 1948 season. They found from 1.0 to 1.7 p.p.m. of DDT on unwashed Delicious that had received one and two cover sprays, and 2.7 to 3.3 p.p.m. on unwashed Winesap apples that had received three cover sprays. Westlake and Fahey also reported from 0.02 to 0.07 p.p.m. parathion on these apples.

A check of the 1949 spray programs of 100 orchardists revealed that 25% applied one DDT cover spray, 47% applied two, 24% applied three, and 4% applied four DDT cover sprays. A high percentage used 1 pound of actual DDT per 100 gallons of water in all cover sprays. Only 15% of these 100 orchardists had added parathion to their cover spray program, and most of the parathion used was applied in the second and/or third cover.

During the 1949 season, chemists at the State College of Washington Tree Fruit Experiment Station, Wenatchee, Washington (Walker 1950), analyzed many unwashed apples for DDT and parathion residue. They found an average of 2.0 p.p.m. of DDT on Delicious, 2.7 on Golden Delicious, 2.5 on Winesap, and 1.7 on Jonathan. The average amount of parathion found was less than 0.03 p.p.m. for all varieties.

By 1951 the picture as far as number of DDT sprays applied per season had not changed much, but in checking the programs of 100 orchardists that year it was noted that more than 50% used dosages, in at least one cover spray, which were less than 1 pound of actual DDT per 100 gallons of water. The use of parathion, however, was on the increase. Of the 100 orchardists, 13% used parathion in one cover spray, 72% used it in two covers, and 15% used it in three cover sprays. Recommendations by the State College of Washington (Anonymous 1951) at that time were: 1 pound of actual DDT per 100 gallons of water in the first cover spray, 0.5 to 1 pound in the second, and 1 pound in the third. Recommendations for parathion suggested dosages from 1.3 to 4 ounces of actual parathion per 100 gallons of water in from one to two cover sprays.

At the end of the 1951 growing season this laboratory collected washed fruit samples for DDT residue determinations from numerous commercial packing warehouses in central Washington. Out of 90 samples, 17 had received one cover spray; 53, two; 17, three; 2, four; and 1, five. The average amount of DDT residue found on the surface of the fruit was 1.2 p.p.m. for Delicious, 2.0 for Golden Delicious, 2.8 for Winesap, and 1.2 for Jonathan.

This laboratory also analyzed fruits grown in this area during the 1954 season and found an average residue of 1.6 p.p.m. of DDT on Winesap. No difference was found in residue level between washed and unwashed samples.

The results of studies of DDT residues on apples grown in central Washington for 1948, 1949, 1951, and 1954 are summarized in table 1.

By 1957–58 more orchardists were finding it necessary to apply four cover sprays and, especially during 1958, a few growers applied five sprays containing DDT. Many of these sprays were applied near to or even beyond the 30-day deadline prior to harvest. By 1958 most orchardists were applying all sprays of DDT at 1 pound actual per 100 gallons of water, and many were also using parathion in practically every cover spray at the rate of 0.25 pound actual per 100 gallons of water. Recommendations allowed application of parathion within 14 days of harvest.

In a study carried out during 1954–56 by Hayes and co-workers (1958), it was shown that no progression in the storage of DDT in the general population of the United States has occurred since this storage was first measured in 1950. This finding indicates that the DDT content of the total diet was not significantly increased during this period.

The purpose of the present study was to determine the average amount of DDT and parathion residues on apples sprayed during the 1958 season in central Washington, and to compare these residues with those determined in past years when fewer sprays, earlier sprays, and lower dosages were used. A study was also made of the effect on residue of a few factors operative during the growth season. The factors studied included formulation, dosage, number of sprays applied, and date of last application.

In addition to the factors listed above, apple packers and processors, as well as growers, have exhibited considerable interest regarding the efficiency of warehouse washing procedures in removing residue. Most apple packing warehouses in central Washington have at least one machine that can be equipped with brushes for the purpose of more thoroughly cleaning the fruit. The water-washing process, whether with or without brushes, is effective in removing visible residue. However, the effectiveness of these procedures in removing oil-soluble residues such as DDT has needed further exploration.

Table 1.—DDT residues (p.p.m.) found at harvest on washed and unwashed apples grown in central Washington during the crop years 1947–58, as reported by various investigators.

Variety	Value	Investigator and Crop Year						
		Westlake & Fahey 1948	Walker 1949	U. S. Public Health Service				
				1951	1954		1958	
		Unwashed	Unwashed	Washed	Unwashed	Washed	Unwashed	Washed
Delicious[a]	No. of samples	5	331	55	—	—	88	44
	Range	1.0–1.7	0.0–7.9	0.2–4.0	—	—	0.3–12.3	0.3–7.0
	Mean ± S.E.[b]	1.3±0.1	2.0	1.2±0.1	—	—	3.4± 0.6	2.3±0.1
Golden Delicious	No. of samples	—	36	8	—	—	10	7
	Range	—	0.0–5.5	0.2–4.6	—	—	0.5–6.3	0.3–3.0
	Mean ± S.E.	—	2.7	2.0±0.4	—	—	3.1± 0.7	1.5±0.5
Winesap	No. of samples	2	294	32	14	21	8	8
	Range	2.7–3.3	0.0–11.9	0.1–5.1	1.0–3.0	1.0–4.0	0.4–12.4	0.5–8.2
	Mean ± S.E.	3.0±0.3	2.5	2.8±0.2	1.6±0.1	1.6±0.2	3.4± 1.2	3.9±0.8
Jonathan	No. of samples	—	129	2	—	—	3	5
	Range	—	0.0–3.3	1.0–1.3	—	—	1.6–7.6	0.7–3.7
	Mean ± S.E.	—	1.7	1.2±0.1	—	—	5.1± 1.8	1.5±0.6

[a] Red and Standard varieties.
[b] Standard error of the mean.

Robinson (1947) reported that washing apples in water immediately after harvest removed about 30% of the residual DDT. Walker (1948) reported that none of the washing treatments he tested effectively reduced high DDT residues. Walker (1949) further noted that when the original DDT residues on apples exceeded 10 p.p.m., the standard washing practices in north central Washington were not sufficient to consistently reduce the residual deposit below the 7 p.p.m. tolerance level.

For these reasons, evaluation of the efficiency of presently used warehouse washing procedures was included in this study.

METHODS AND MATERIALS.—Apple samples for the 1958 residue study were collected from commercial packing warehouses. Most of the Delicious and Jonathan samples were taken within 2 weeks after harvest and none was in storage longer than 8 weeks. Although a few Golden Delicious and Winesap samples were taken soon after harvest, most of these two varieties were taken after they had been in storage 4 to 8 weeks. Unwashed samples were taken just prior to their entrance into the washer, and washed samples were taken from the grader bins after they had been water washed, sorted, and sized by weight preparatory to packing for shipment to market. The apples ranged from sizes 125 to 138 (2 14/16 to 2 13/16 inches in diameter). Sufficient firm fruit to completely fill a 1-gallon, wide-mouthed glass jar was washed with 750 ml. of hexane (Skellysolve B) for 15 minutes in an end-over-end type tumbling machine, and aliquots of the hexane extract were taken for analysis. Determinations of DDT residues were carried out by the method of Schechter *et al.* (1945), as modified by Mattson *et al.* (1953). Parathion analyses were made by the method of Averell & Norris (1948).

RESULTS.—*DDT Residues.*—A total of 123 samples of apples was analyzed for DDT residue. These samples had received from one to five DDT cover sprays. DDT found on the fruit ranged from 0.3 to 12.4 p.p.m., with an average of 3.0 p.p.m. on all samples, both washed and unwashed. Unwashed samples averaged 3.7 p.p.m. and washed samples averaged 2.4 p.p.m.

Table 1 shows the amount of DDT found on each of the four varieties of apples, unwashed and washed. As has been reported by other workers, slightly higher residues were found on Winesap than on the other varieties. For each variety and for all samples considered together, the unwashed apples had a significantly higher residue than the washed samples. Only nine unwashed (15.2%) and one washed sample (1.6%) had residues above the 7 p.p.m. tolerance level. As indicated in table 1, the DDT residue values for the 1958 crop are somewhat higher than in previous years for which similar studies are available.

Table 2 shows the relationship of the number of cover sprays applied during the season to the amount of DDT residue. The data presented show that with an increase in the number of cover sprays applied, a corresponding increase in the amount of residue is found on the fruit. It is interesting to note that, of the 12 samples that received five cover sprays, five (42%) exceeded the tolerance level. Even the mean value for this group of samples

Table 2.—Relation of number of cover sprays applied to apples to the amount of DDT residue found at harvest.

No. of Cover Sprays	Average Interval Between Last Spray and Harvest (Days)	No. of Samples	Residue (P.P.M.)	
			Range	Mean ± S.E.[a]
1	60.0	2	0.9 to 0.9	0.9±0.0
2	51.1	14	.3 to 3.7	1.5±0.3
3	38.9	61	.3 to 8.2	2.6±0.3
4	23.9	34	1.1 to 7.6	3.3±0.3
5	17.2	12	1.1 to 12.4	6.4±1.0

[a] Standard error of the mean.

Table 3.—Effect of warehouse washing procedures on DDT residue on apples.[a]

Residue Level	DDT Residue (p.p.m.) After Indicated Treatment		
	Unwashed	Washed, No Brushes	Washed, Brushes
High			
Range	9.0–12.3	2.7–3.1	3.4–4.9
Mean ± S.E.[b]	10.6± 1.0	2.8±0.1	4.0±0.5
Medium			
Range	5.4– 6.2	2.0–3.7	2.6–4.5
Mean ± S.E.	5.9± 0.3	2.7±0.5	3.8±0.6
Low			
Range	2.0– 2.6	1.1–1.3	1.2–1.4
Mean ± S.E.	2.4± 0.2	1.2±0.1	1.3±0.1
Mean			
Range	2.0–12.3	1.1–3.7	1.2–4.9
Mean ± S.E.	6.3± 1.0	2.2±0.3	2.9±0.5

[a] Each mean represents the average for three replicate samples.
[b] Standard error of the mean.

(6.4 ± 1.0 p.p.m.) approaches the 7 p.p.m. figure. These results suggest that the application of more than five cover sprays may give residue values of such magnitude that virtually all samples may exceed the tolerance level.

The mean time intervals between last spray and harvest for samples having different numbers of cover sprays are also shown in table 2. It is apparent that a negative correlation exists between interval from last spray to harvest and DDT residue level. However, the number of cover sprays and time interval between last spray and harvest are also interrelated, so that, in general, as the number of cover sprays is increased, the interval between last spray and harvest will become shorter. In order to separate the relative magnitude of the effects of these two interrelated factors on residue level, statistical analysis involving determination of the partial correlation coefficients for the effect of number of cover sprays and of interval between last spray and harvest on residue level was carried out by the method described by Snedecor (1956). The effects of both number of cover sprays and of interval between last spray and harvest on residue level was found to be significant at greater than the 99% level. The correlation coefficient for residue level with number of sprays (+0.473) was somewhat greater than that with interval between last spray and harvest (−0.403).

An additional factor that might be expected to influence the residue level at harvest is the formulation used, particularly the presence or absence of a spreader or sticker-type adjuvant in the spray mix. Little information was obtained on this point, however, since only four of the apple samples tested had been sprayed with formulations containing a spreader or sticker. The material used on the test samples was X-77, a combined spreader-activator, manufactured by Colloidal Products Corporation, Sausalito, California. The principal functioning agents in this material included alkyl arylpolyoxyethylene glycol, isopropanol, and free fatty acids. Two to four ounces were used per 100 gallons of spray mixture. These four samples (two Delicious and two Winesap) had residues before washing of from 1.3 to 5.8 p.p.m. with a mean of 3.2 p.p.m.

Effect of Washing Equipment.—In order to define more clearly the effectiveness of various types of washing equipment in DDT residue removal, an experiment was conducted to compare the residues on unwashed fruit, on fruit that had been water-washed in a machine without brushes, and on fruit washed in a machine equipped with brushes. Fruit samples from lots where the approximate residue level was known were used to set up a replicated experiment, including samples of relatively high, medium, and low residues, respectively. Amounts of DDT were determined on three replicate samples of apples from each of the three sources, including samples unwashed, washed without brushes, and washed with brushes. The whole sampling procedure was carried out in a commercial packing warehouse and typical commercial washing machines were used. Results are shown in table 3.

On the basis of the data presented, it appears that the washing process removes significant amounts of DDT, especially where higher residues are present before washing. It will be noted that DDT residue on samples containing as high as 12.3 p.p.m. was reduced to levels well below the 7 p.p.m. tolerance when the fruit was run through the washer. It should be emphasized, however, that not all samples having prewash residues in this range will be converted to a sub-tolerance level by washing. One sample (from table 1), for example, was reduced only from 12.4 p.p.m. to 8.2 p.p.m. The 37 other samples that were analyzed both before and after washing showed an average DDT residue of 3.2 p.p.m. before washing and 1.5 p.p.m. after washing.

It is of interest to note that, in the replicated washing experiment, the machine without brushes appeared to be more efficient in removing residue than the machine equipped with brushes. One might speculate that, over a period of time during the early part of the packing season, the brushes would accumulate a deposit of DDT. Once DDT is on the brushes, their action might no longer be to remove DDT from all the fruit, but rather to remove some DDT from fruits with extra heavy deposits and to add small amounts of DDT to fruits having little or no DDT on their surface prior to entering the washer.

Parathion Residues.—A total of 24 unwashed apple samples was analyzed for parathion residue. The varieties sampled included Delicious, Jonathan, and Winesap, which had received parathion in from one to five cover sprays. The amount of parathion found on the fruit ranged from 0.01 to 0.32 p.p.m. with an average of 0.10 p.p.m. The amount found on Delicious averaged 0.09 p.p.m.; on Jonathan, 0.08 p.p.m.; and on Winesap, 0.24 p.p.m. These values are somewhat higher than the 0.02 to 0.07 p.p.m. found by Westlake & Fahey (1950) on 1948 fruit and much higher than the <0.03 p.p.m. average reported by Washington State College Tree Fruit Experiment Station chemists for 1949 fruit (Walker 1950).

No correlation was found between the amount of parathion residue at harvest and the number of parathion sprays applied. The average residue for samples that had received three sprays was 0.11 p.p.m. and for samples that had been sprayed four times was 0.10 p.p.m.

There was a definite correlation between the amount of parathion residue and closeness of spray to harvest. At harvest time on September 15 no parathion residue could be found on apples to which the last parathion spray was applied during May. Where the last spray was applied

during June or July, an average of 0.02 p.p.m. was found. The average amount of parathion found on apples sprayed during August and September was 0.02 p.p.m. for those sprayed 25 to 27 days prior to harvest, 0.12 p.p.m. for those sprayed 18 to 21 days before harvest, and 0.12 p.p.m. for those sprayed only 10 to 15 days before harvest.

All the parathion residue values for apples from the 1958 crop were well below the 1 p.p.m. tolerance level.

DISCUSSION.—Although residues of both DDT and parathion on apples sprayed during the 1958 season in central Washington were somewhat higher than those found in similar studies carried out between 1948 and 1954, the increase in amount of residue resulting from increased dosage and additional, later cover sprays has not been great. In almost all instances, the residues have been below the tolerance level. The small, definite increase noted in residue level for both DDT and parathion over the 10-year period covered, however, emphasizes the need for giving close attention to spray programs in the future. As insect resistance or other factors force more intensive application of DDT and parathion, additional surveys of the effect on residue levels should be carried out.

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STUDIES ON SOME POTENTIAL HEALTH HAZARDS ASSOCIATED WITH INSECTICIDE RESIDUES ON POME FRUITS*

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The most spectacular manifestation of the toxicity of modern pesticides is the acute toxic effect which some of the more hazardous compounds may have on severely-exposed individuals. The death or serious illness of a sprayman due to over-exposure to a spray material represents an incident which causes all of us in the fruit industry to stop and think. Even more impressive is the accident in which a child is poisoned by playing with an "empty" insecticide container or by eating or drinking a pesticidal formulation from an unlabelled container.

SPRAY POISONING

Let us, for a moment, review the situation with regard to acute pesticide poisoning as we have seen it over the past two spray seasons in Central Washington.

During the 1958 spray season there occurred a rather large number of illnesses allegedly due to pesticides in Central Washington (1). These cases included two fatalities and approximately 100 illnesses ranging in severity from mild (with no time lost from work) to serious (involving several days of hospitalization). In 71 of the 100 cases, it was considered that an etiology of pesticide poisoning was established by exposure history, clinical symptoms, and or laboratory tests. The incidence of poisoning was somewhat lower during 1959. Most of the poisoning cases involved parathion, the most widely used of the more toxic pesticides in this area. However, phosdrin and demeton (Systox) were also involved. No cases of chlorinated hydrocarbon poisoning were seen.

STUDIES ON RESIDUES

Although admittedly less spectacular, a much more inclusive aspect of the potential hazard of insecticides involves residues on food. This problem is of importance to the entire population of this country who will be eating harvested produce bearing small quantities of pesticide residues.

There have been no cases of illness in this country from insecticide residues on food when pesticidal formulations have been used according to directions (2). The same is true in Britian (3). However, there have been several instances in which insecticides used quite improperly on foods have led to poisoning (4), (5).

The primary toxicological problem associated with residues, however, is not the possibility of a single exposure producing illness, but rather the possible adverse effect on health of consumption of small amounts of these chemicals over a period of years.

Residues on Apples: Our laboratory has recently reported a study of the trend over the past ten years of DDT and parathion residues on apples grown in Central Washington (6). Since the first general use of DDT and parathion for insect and mite control on tree fruits in Central Washington during 1947-48, the amount of these insecticides used per season to maintain adequate control has increased gradually. Although there has been a small increase in the dosage rate of DDT, the major changes contributing to the residue problem are the application during the last few years of a greater number of cover sprays containing DDT during the growing season and the fact that the last cover is often applied closer to harvest. The dosage rate and number of parathion sprays have also increased.

A total of 123 samples of apples from the 1958 crop in Central Washington was analyzed for DDT residue. These samples had received from one to five DDT cover sprays. The amounts of DDT found on the fruit ranged from 0.3 to 12.4 ppm with an average of 3.0 ppm on all samples, both washed and unwashed. Unwashed samples averaged 3.7 ppm and washed samples averaged 2.4 ppm. Only 1 washed sample (1.6 percent) had a residue above the 7 ppm tolerance level.

Either increasing the number of cover sprays or decreasing the time interval between last spray and harvest caused an increase in DDT residue level. The conventional warehouse washing opera-

* From the Communicable Disease Center, Bureau of State Services Public Health Service, U. S. Department of Health, Education, and Welfare, Wenatchee, Washington.

tion definitely lowered the residue content, but there appeared to be little or no difference between washing with or without brushes in the machines.

A total of 24 unwashed apple samples was analyzed for parathion residues. The amount of parathion found ranged from 0.01 to 0.32 ppm with an average of 0.10 ppm. These values, even though they were determined on unwashed fruit, are well below the 1 ppm tolerance level for parathion residue on apples.

Although residues of both DDT and parathion on apples sprayed during the 1958 season in Central Washington were found to be somewhat higher than those in similar studies carried out between 1948 and 1954, the increase in amount of residue resulting from increased dosage and additional, later cover sprays has not been great. The small, definite increase noted in residue level for both DDT and parathion over the 10-year period covered, however, emphasized the need for giving close consideration to spray programs in the future.

Residues on Pears: The chlorinated hydrocarbon insecticide that has been most widely used on pears in Central Washington during the past few years is DDT. DDT residues at harvest time have generally been well under the 7 ppm tolerance level. During the 1957 season two additional chlorinated hydrocarbon compounds, dieldrin and toxaphene, were used in an effort to control the insect, pear psylla, which had become increasingly difficult to control with parathion.

Our laboratory carried out a study to determine the amounts of DDT, dieldrin, and toxaphene residues on pears sprayed during the 1957 season.

A total of 69 unwashed samples of pears was analyzed for DDT residue. The samples had received from one to three cover sprays containing DDT. The residue found ranged from 0.1 to 3.7 ppm, with a mean of 1.2 ppm.

The dieldrin residue was determined for 60 samples of pears. These samples had received one or two cover sprays containing dieldrin. The dieldrin residue ranged from 0.05 to 0.21 ppm, with a mean of 0.07 ppm.

A total of 16 pear samples was analyzed for toxaphene residue. All had received only one toxaphene spray. The residues ranged from 0 to 1.90 ppm, with a mean of 0.53 pm.

Of the total of 145 analyses summarized above, no residue for a single insecticide was found to be above the tolerance level.

Combined Residues: Forty-five of these pear samples had received both DDT and dieldrin sprays and ten samples had received DDT, dieldrin and toxaphene. The usage of two or more of these related chemicals on a single crop raises the possibility of exceeding the allowable tolerance for the combination of these materials. The U.S. Food and Drug Administration regulations state that, where two or more pesticides with similar pharmacologic actions are present, the combined percentile residues shall not exceed one hundred (7).

The combined percentile residues of these samples have been calculated and the results are shown in tables 1 and 2. One of the samples (2.2 percent) sprayed with DDT and dieldrin and two of the samples (10 percent) sprayed with DDT, dieldrin, and toxaphene exceeded the allowable combined tolerance. In all three instances in which the combined residues exceeded the tolerance, a relatively high dieldrin residue (80 percent or above) was the major contributing factor.

It should be noted that these values apply to unwashed fruit. It may be that washing would reduce some or all of these amounts below the tolerance level. However, the fact that these samples were near or above the tolerance level points out the necessity for the orchardist giving attention to the mixed residue regulation when planning his spray program.

Additional mixed residue studies on the 1959 apple crop for kelthane-DDT, guthion-sevin, and parathion-sevin are being carried out. (Although sevin is not an organic phosphorus material, it is, due to its anticholinesterase activity, considered for purposes of mixed residue determination with the organic phosphorus insecticides.) Although all these analyses have not yet been completed, it does not appear at this time that any of the above combinations present a significant residue problem when used according to recommended practices.

DDT in Apple Pomace: Another problem of considerable interest involves DDT residue in apple pomace. It has been common practice for apple processing plants, engaged in vinegar or juice production, to dispose of waste apple pulp by offering it for sale, mainly as feed for livestock. The pomace is apparently a good food source and it has been

reported that young heifers fed on apple pomace gained weight significantly faster than similar animals fed their usual ration of sorge-Sudangrass-soybean silage and cottonseed cake (8). The dried pomace has also been used as a feed for poultry and rabbits and as a bait material in certain insecticidal formulations.

The processing plants utilize either whole, cull apples or residual cores and peelings. Results of determination of DDT residues at harvest on unwashed and washed apples from the 1958 crop in Central Washington are given above. These figures apply to whole apples. The values for cores and peelings only would undoubtedly be higher, since essentially all the residue is on the surface of the apple.

It is well known that DDT accumulates in the fatty tissues of animals fed feed contaminated with DDT and it has been shown also that dietary DDT is excreted in the milk of lactating animals. Thomas **et al.** (9), found that in calves the DDT concentration in the fat was proportional to the intake. Radeloff and his coworkers (10) reported that yearling Herefords fed 10 ppm of DDT in all items of their diet for 30 days showed 6.8 ppm in fat on the last day of feeding. Fahey and Brindley (11) found that Hereford steers, grazing on corn stover containing DDT up to 23.3 ppm, showed 1.0 to 2.8 ppm of DDT in fat at the end of one month and 5 ppm after two months. After taking the animals off the DDT-contaminated diet, their stored DDT diminished quite rapidly. Within three months only 0.1 ppm or less was found.

In the present study, samples of dry pomace, wet pomace, and apple juice were taken for analysis for DDT content on three occasions at weekly intervals. On each occasion three replicate samples were taken of each product. The processing plant, at the time of the study, was utilizing only the cores and peelings of Delicious apples. The amount of DDT found in the nine dry pomace samples ranged from 32 to 46 ppm with an average of 36 ppm. From 6 to 18 ppm (average 11 ppm) was found in the wet pomace. No DDT was detected in the juice.

On the basis of these results it appears that feeding livestock on apple pomace might result in concentrations of DDT in milk and meat above their respective tolerance limits of 0 and 7 ppm. This danger has been noted by the U. S. Food and Drug Administration which has recently ruled that apple pomace containing any amount of DDT is unsuitable as a feed for dairy animals and that care should be taken in feeding apple pomace containing DDT to meat animals to be sure that the meat from such animals does not contain residues in excess of the tolerance (12).

Our laboratory has given some attention to methods for decreasing the DDT content of pomace. Although apples destined for pomace could be subjected to

Table 1. Combined Residues on Pears Sprayed with DDT and Dieldrin, Central Washington, 1957 (45 samples).

Material	Residue	
	ppm	Percent of Tolerance
DDT	1.11 (0.11 to 3.70)	16 (2 to 53)
Dieldrin	0.07 (0.05 to 0.21)	28 (20 to 80)
Total		44 (21 to 110)

Table 2. Combined Residues on Pears Sprayed with DDT, Dieldrin and Toxaphene, Central Washington, 1957 (10 samples).

Material	Residue	
	ppm	Percent of Tolerance
DDT	1.80 (0.25 to 3.56)	26 (3 to 51)
Dieldrin	0.10 (0.05 to 0.21)	40 (20 to 84)
Toxaphene	0.65 (0.25 to 1.30)	9 (3 to 27)
Total		75 (36 to 141)

much more severe washing procedures than fruit for the fresh market, these additional procedures would apparently not be economically feasible. Another approach which is being investigated is fermentation. Other workers have reported a decrease in DDT residue during the fermentation process of ensilage production (13), (14).

SUMMARY

Studies of the trend of DDT and parathion residues on apples grown in Central Washington between 1948 and 1958 have shown a small but definite increase in the residue of both these insecticides, associated with increased dosage and additional, later cover sprays. In 145 samples of pears from the 1957 crop, treated with DDT, dieldrin, or toxaphene, no residue for a single insecticide was found to be above the tolerance level. However, one sample (of 45 tested) treated with both DDT and dieldrin and two samples (of 10 tested) treated with DDT, dieldrin, and toxaphene were above the allowable combined tolerance level. DDT residue levels averaged 36 ppm in dry apple pomace and 11 ppm in wet pomace.

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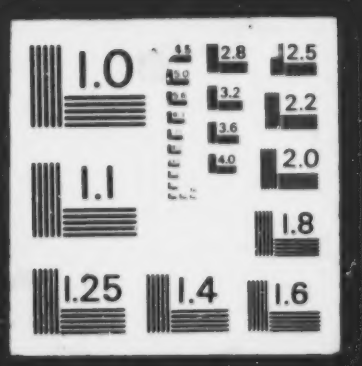

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RESIDUES OF SOME PESTICIDES FOUND ON APPLES GROWN IN CENTRAL WASHINGTON*

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The U. S. Public Health Service, at the Wenatchee Field Station, has been studying the potential health hazards associated with the intensive use of various pesticides in agriculture. These studies include both measurement of hazard to workers and determination of the hazard to the general population through incidental exposure or consumption of the compounds on harvested crops.

The primary toxicological problem associated with residues is not the possibility that a single exposure will produce illness, but rather the possible adverse effect on health of consumption of small amounts of these chemicals over a period of years.

Our main interest in measuring residues of pesticides on tree fruits in Washington is to follow the trend of residues from year to year. For example, through checking spray programs and analyzing for residues we may determine that the amount of DDT or other pesticide that the general public is consuming by eating Washington apples may be greater during certain years than during others.

Fieldmen and orchardists have expressed an interest in knowing just how much residue of certain pesticides is present on fruit at packing time. Although we all have been informed that residues should be within allowable tolerances if applications are made according to recommendations, the orchardist, no doubt, sometimes wonders just how much residue of certain pesticides results from his spray program. This is especially true if he has found it necessary to apply more sprays than usual. Is he producing residues very near the allowable tolerance or is there considerable leeway?

Individual Residues

Results of 439 analyses of residues found on Delicious and Winesap apples from 1958 through 1960 are shown in table 1. Analyses for parathion and Guthion were made according to the method of Averell and Norris (1), Sevin analyses were by the method of the Union Carbide Chemical Company (5), DDT was analyzed by the Schechter-Haller method (3), Kelthane by a modification of the Stiff-Castillo method (4), and Tedion by the method of Fullmer and Cassil (2). Some of this series of samples were collected to obtain fruit that, because of the particular spray program involved would be suspected of having high residues. If these samples were below the allowable tolerance, we could then feel that all others would also probably fall below the tolerance. These data for parathion, Guthion, Sevin, DDT, Kelthane, and Tedion show that the residues found on all samples ranged well below the individual allowable tolerances, except for some DDT samples during the 1958 season. In the case of the 1958 DDT data, out of seven samples that were

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TABLE 1

Residues of some pesticides found on both washed and unwashed Delicious and Winesap apples grown in Central Washington.

Compound	Allowable Tolerance (PPM)	Year	Samples	Residue (PPM) Range	Average
Parathion	1	1958	24	0.01 to 0.32	0.10
Guthion	2	1959	26	0.00 to 0.60	0.10
Sevin	10	1958-59	26	0.00 to 1.00	0.10
DDT	7	1958	123	0.30 to 12.40[1]	3.00
DDT	7	1959	19	0.40 to 5.40	2.40
DDT	7	1960	14	0.00 to 0.30	0.12
Kelthane	5	1958	59	0.20 to 2.60	0.90
"	5	1959	52	0.20 to 1.20	0.60
Tedion	5	1960	60	0.10 to 2.10	0.34

[1] Of seven samples that were above tolerance before washing, only two remained above tolerance after washing, and these had been sprayed within 30 days of harvest.

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over the 7-ppm tolerance before washing, only two remained above tolerance after having passed through the warehouse washing machine. Also, all of these seven involved spray applications that were closer to harvest than the 30-day period recommended by the U. S. Department of Agriculture. The highest amount of DDT residue found on unwashed apples that were sprayed over 30 days prior to harvest was 6.3 ppm. The relationship of the number of cover sprays that these 1958 DDT samples had received during the season to the amount of residue found is shown in table 2. As the number of cover sprays was increased, the interval between the last spray and harvest became shorter and the residue greater.

Combined Residues

Another facet of the residue problem concerns the use of two or more similar compounds on the same crop, raising the possibility of exceeding the allowable tolerance for the combination of these materials. The U. S. Food and Drug Administration regulations state that, where two or more pesticides with similar pharmacologic actions are present, the combined percentile residues shall not exceed one hundred (6). The basic chemical structure and pharmacologic action of DDT, Kelthane, and Tedion are similar; therefore, they are classified together for residue purposes. Also, parathion, Guthion, and Sevin are in a class together. Although Sevin is a carbamate, it is, due to its anticholinesterase activity, classed with the organic phosphorus compounds for purposes of mixed residue determination.

Mixed residues of Guthion and parathion were found to be much below the allowable tolerance limit for combined residues. Analyses of residues on eight samples that had received both Sevin and Guthion sprays showed the combined residues to be from 6 percent to 31 percent of the allowable combined tolerance.

With respect to the chlorinated organic compounds, however, the possibility of exceeding the allowable combined tolerance was found to be greater. This finding may be due primarily to the longer residual life of these compounds. The number of times during the season that the sprays are applied and the number of different compounds of the same class that are applied to the same fruit, also has a direct bearing on the amount of combined residues present. Examples of combined DDT and Kelthane residues on Delicious and Winesaps are shown in table 3. These samples

TABLE 2

Relation of number of cover sprays applied to apples to the amount of DDT residue found at harvest, 1958, Central Washington.

No. of Cover Sprays	Average Interval Between Last Spray and Harvest (Days)	Residue (PPM)	
		Range	Average
1	60.0	0.9 to 0.9	0.9
2	51.1	0.3 to 3.7	1.5
3	38.9	0.3 to 8.2	2.6
4	23.9	1.1 to 7.6	3.3
5	17.2	1.1 to 12.4	6.4

TABLE 3

A few of the higher combined DDT and Kelthane residues found on Delicious and Winesap apples in Central Washington, 1958.

Sample No.	Washed (W) or Unwashed (U)	DDT Residue (7 PPM Tol.)		Kelthane Residue (5 PPM Tol.)		Total %—DDT & Kelthane Combined
		PPM	% of Tol.	PPM	% of Tol.	
1	U	6.2	88	2.4	48	136*
1	W	4.5	64	1.7	34	98
2	U	10.5	150	1.3	26	176*
2	W	3.4	48	0.9	18	66
3	U	6.2	88	2.2	44	132*
3	W	2.6	37	1.1	22	59
4	U	5.4	77	1.7	34	111*
4	W	3.3	47	1.1	22	69
5	U	12.4	177	2.1	42	219*
5	W	8.2	117	2.0	40	157*

* Over combined percentile residue tolerance.

were taken in 1958 which was a year of great insect infestation and spraying was at a high level. If the percentage of allowable residues found in the DDT and Kelthane analyses are added together, the total is above 100 percent for the unwashed samples shown. The fruits therefore contain amounts of pesticides above the allowable combined tolerances. It should also be noted in table 3 that the same samples after washing were all reduced in residue content to a point below the allowable tolerance, with the exception of one sample. This one sample was found to be over tolerance for DDT alone without applying the combined residue rule.

During the 1960 spray season, very few orchards received applications that produced a combined residue of DDT, Kelthane, and Tedion, all of which are classed together for combined residue purposes. A number of them, however, had been sprayed with DDT and Tedion or Kelthane and Tedion. Analyses of residues on 14 samples of apples that had received both DDT and Tedion sprays showed the combined residues to range from 6 to 13 percent of the allowable combined percentile tolerance.

Analyses of 111 Kelthane and 60 Tedion samples have shown the residues of each to average less than 20 percent of their individual tolerances on apples, indicating that combined residues of these two compounds should be no great problem except under unusual circumstances such as the application of both Kelthane and Tedion sprays relatively close to harvest. If the residues for Kelthane and Tedion are the average that have been found, the combined percentile residue would be only 19 percent. However, if the highest values found for both are considered, the percentile residue would be 94 percent. Even though this figure is below the combined tolerance level of 100 percent it is near enough to indicate that some caution should be observed when considering the use of both Kelthane and Tedion sprays relatively close to harvest, and especially so if DDT is also present.

Discussion and Summary

It appears from the data presented here that individual or combined residues of various pesticides including DDT, Kelthane, Tedion, Guthion, parathion, and Sevin on apples in Central Washington in the 1960 season were generally well below the allowable tolerances. No samples were above the tolerance level when sprays were applied according to recommendations.

One must recognize, however, that directions are not always adhered to. Occasionally, orchardists will encounter a difficult insect or mite problem and apply sprays at higher than recommended dosage rates, too close to harvest or apply a greater number of sprays than recommended. In these instances, individual or particularly mixed residues may exceed the tolerance level.

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